

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Patent Application of  
TAKESHI MATSUMOTO ET AL

5    Serial No. 10/535,331                      Group Art Unit: 1755  
     Filed: May 18, 2005                      Examiner: Wood, Elizabeth D  
     Title: EXHAUST GAS PURIFYING CATALYST AND PROCESS FOR  
     PURIFYING OF EXHAUST OF GAS

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DECLARATION  
UNDER 37 C.F.R. 1.132

     I, Takuji Nakane, a citizen of Japan, residing at  
15    344-1-706, Waku, Aboshi-ku, Himeji-shi, Hyogo-ken, Japan,  
     declare:

     I. I am one of co-inventors in the above referenced  
     application, and a chemist as well as a Researcher of AC  
     Research Laboratory of ICT CO., LTD., one of co-assignees  
20    of this application, on the subject matters relating to this  
     application.

     I graduated from Tokyo Institute of Technology, Faculty  
     of Science, Department of Chemistry in March 1997 and Graduate  
     School of Tokyo Institute of Technology, Department of  
25    Chemistry in March 1999, and obtained a master degree majoring  
     surface science.

     Since April, 1999, I have been the employee of ICT CO.,  
     LTD. at AC Research Laboratory and have been engaged in the  
     research work with respect to catalysts for purifying exhaust  
30    gas of diesel engine.

     I am well acquainted with all the other co-inventors  
     in this case, having worked with them on the development of

the invention described in this application.

II. In order to compare the effects of the catalysts of this application with those catalysts of Labarge et al I (US 6,489,259) and Labarge et al II (US 2002/0086793), I have conducted the following experiments:

#### Example 8

A catalyst was prepared by a similar method to Example 4 of this application. During the test for performance of purifying exhaust gas, light oil containing 0.03% by weight of sulfur was used as a fuel for an internal combustion engine. The results were summarized in Table 1A.

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TABLE 1A

Inlet temp. of catalyst layer	HC (ppm)	CO (ppm)	NOx (ppm)	O <sub>2</sub> (vol.%)	CO <sub>2</sub> (vol.%)	SO <sub>2</sub> (ppm)	L.O. (mL/min)
500°C	30	90	380	6.5	10.2	12	8.5
450°C	45	100	360	8.1	9.1	-	7.0
400°C	50	110	320	9.6	8.0	-	6.5
350°C	60	120	240	11.0	7.0	-	5.5

L.O.: Light oil

#### Comparative Example 3

A similar method to Example 4 of this application was carried out except that ZSM-5 having 2-4  $\mu$ m of particle diameter was used. Test results of Comparative Example 3 and Example 8 were summarized in Table 5.

TABLE 5 Initial NOx purification activity (%)

	500°C	450°C	400°C	350°C
Example 8	45	42	43	23
Comparative Example 3	35	35	37	22

## Example 9

Catalyst was prepared by a similar method to Example  
 5 4 of this application except that 116g of ZSM-5, 91g of  
 $\beta$ -zeolite, 42g of Copper nitrate and 42g of silica sol were  
 used. In this case, weight ratio of ZSM-5 and  $\beta$ -zeolite was  
 11:8. During the test for performance of purifying exhaust  
 10 gas, light oil containing 0.003% by weight of sulfur as a  
 fuel for an internal combustion engine. The results were  
 summarized in Table 1B.

TABLE 1B

Inlet temp. of catalyst layer	HC (ppm)	CO (ppm)	NOx (ppm)	O <sub>2</sub> (vol.%)	CO <sub>2</sub> (vol.%)	SO <sub>2</sub> (ppm)	L.O. (mL/min)
500°C	200	170	330	6.5	10.2	15	8.5
450°C	160	160	300	8.1	9.1	-	7.0
400°C	140	180	250	9.6	8.0	-	6.5
350°C	180	240	200	11.0	7.0	-	5.5

L.O.: Light oil

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## Example 10

A similar method to Example 9 described above was carried  
 out except that 160g of ZSM-5 and 43g of  $\beta$ -zeolite were used.  
 In this case, weight ratio of ZSM-5 and  $\beta$ -zeolite was 5:1.

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#### Comparative Example 4

A catalyst was prepared by a similar method to Comparative Example 1, and a similar purification test was carried out.

The results of the test were shown in Table 2A including  
5 those of Examples 9 and 10 and Comparative Example 4.

TABLE 2A

	CuO	ZSM5 (MFI zeolite)		BEA ( $\beta$ zeolite)	ZSM-5/ $\beta$ -zeolite
		$\text{SiO}_2/\text{Si}_2\text{O}_3 = 70$	$\text{SiO}_2/\text{Si}_2\text{O}_3 = 30$		
		Average Crystal: less than $0.05\mu\text{m}$	Average Crystal: $0.4\mu\text{m}$		
Example 9	7	55		40	11:8
Example 10	7	76		19	5:1
Comparative Example 4	7	95			1:0

In Table 2: Unit: g/ litter catalyst

TABLE 6 Initial NOx purification activity (%)

	500°C	450°C	400°C	350°C
Example 9	44	45	44	33
Example 10	43	45	47	39
Comparative Example 4	42	46	47	28

TABLE 7 NOx purification activity (%) after durability test

	500°C	450°C	400°C	350°C
Example 9	42	43	41	20
Example 10	44	42	40	23
Comparative Example 4	43	39	40	15

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### III. CONCLUSION

According to Table 5, a catalyst of this invention (i.e., the catalyst of Example 8), in which ZSM-5 had an average particle diameter not greater than 0.5  $\mu\text{m}$ , was more active in reducing NOx in exhaust gas than a comparative catalyst (i.e., the catalyst of Comparative Example 3), in which ZSM-5 had an average particle diameter greater than 0.5  $\mu\text{m}$ .

According Tables 6 and 7, a catalyst of this invention (i.e., the catalyst of Example 9 or 10), in which the weight ratio of ZSM-5 to  $\beta$  zeolite fell within the range of 1:0.1 to 1:5 recited in claim 1, was more active in reducing NOx in exhaust gas at low inlet temperature, i.e., 350°C, than a comparative catalyst (i.e., the catalyst of Comparative Example 4), which, like LaBarge's catalyst, contained ZSM-5, but not  $\beta$  zeolite.

The undersigned Takuji Nakane declare that all the statements made herein are true; and further that these statements are made with the knowledge that willful false  
5 statements and the like so made are punishable by fine or imprisonment, or both under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

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Dated this 18th day of February, 2008.

By Takuji Nakane  
Takuji Nakane

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